# Boltzmann Equations for Spin and Charge Relaxations in Superconductors

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In a superconductor coupled with a ferromagnetic metal, spin and charge imbalances can be induced by injecting spin-polarized electron current from the ferromagnetic metal. We theoretically study a nonequilibrium distribution of quasiparticles in the presence of spin and charge imbalances. We show that four distribution functions are needed to characterize such a nonequilibrium situation, and derive a set of linearized Boltzmann equations for them by extending the argument by Schmid and Schön based on the quasiclassical Green's function method. Using the Boltzmann equations, we analyze the spin imbalance in a thin superconducting wire weakly coupled with a ferromagnetic electrode. The spin imbalance induces a shift $\delta\mu$ ($-\delta\mu$) of the chemical potential for up-spin (down-spin) quasiparticles. We discuss how $\delta\mu$ is relaxed by spin-orbit impurity scattering.

KEYWORDS: nonequilibrium superconductivity, spin imbalance, charge imbalance, spin-orbit interaction

## 1. Introduction

Electron current injected into a superconductor produces a nonequilibrium distribution of quasiparticles.[1–9] A number difference between electrons and holes arises in such a nonequilibrium situation.[4,5] This is called charge imbalance. The charge imbalance induces an excess quasiparticle current, which results in a potential difference between pairs and quasiparticles.[1,4,5] Although early experiments on the charge imbalance have focused on spin-independent phenomena, we can now study spin-related ones by injecting spin-polarized electron current into superconductors by using a ferromagnetic metal as an injection electrode.[10] If injected electron current is spin polarized, there arises spin imbalance between up- and down-spin quasiparticles in addition to the charge imbalance. In this case, a potential difference between up- and down-spin quasiparticles arises in addition to a spin-independent potential difference due to the charge imbalance. The spin imbalance in mesoscopic superconductors has recently attracted much attention both experimentally[11–13] and theoretically.[14,15] The main attention is focused on its relaxation due to spin-flip processes.

The most general framework to understand the charge imbalance in superconductors is presented by Schmid and Schön.[7] Based on kinetic equations for the quasiclassical Green's functions, they showed that nonequilibrium quasiparticle distributions are described by two

distribution functions $f_{\rm T}(\boldsymbol{r},\epsilon)$ and $f_{\rm L}(\boldsymbol{r},\epsilon)$, where $\epsilon$ represents quasiparticle energy measured from the chemical potential $\mu$ at equilibrium, and derived a set of linearized Boltzmann equations for them. It has turned out that $f_{\rm T}$ and $f_{\rm L}$ describe the charge imbalance and the related excess quasiparticle energy, respectively, and that they are coupled with respectively transverse and longitudinal variations of the pair potential. By using the Boltzmann equations, we can describe the charge imbalance including effects of inelastic phonon scattering. However, their framework is restricted to spin-independent phenomena, and cannot apply to the case where the spin imbalance plays a role. To fully understand nonequilibrium quasiparticle distributions in the presence of a spin-polarized current injection, we need a framework by which we can describe both the spin and charge imbalances in a unified manner including effects of phonon scattering. Although a few theoretical treatments for the spin imbalance in superconductors have been reported so far,[14,15] they do not satisfy all our requirements.

In this paper, we study a nonequilibrium quasiparticle distribution in the presence of the spin and charge imbalances. To describe such a nonequilibrium situation, we adopt the kinetic equation approach by Schmid and Schön. We introduce four distribution functions $f_{\rm L+}$, $f_{\rm L-}$, $f_{\rm T+}$ and $f_{\rm T-}$ to characterize quasiparticle distributions. It is shown that $f_{\rm L+}$ and $f_{\rm T-}$ represent the spin and charge imbalances, respectively, and $f_{\rm L-}$ and $f_{\rm T+}$ represent the excess quasiparticle energy and the energy imbalance between up- and down-spin quasiparticles, respectively. The suppression of the pair potential due to a current injection is determined by $f_{\rm L-}$. We derive a set of linearized Boltzmann equations for the distribution functions in steady states assuming that the spin imbalance is relaxed by spin-orbit impurity scattering. We show that the four distribution functions are decoupled with each other in the resulting Boltzmann equations. This indicates that we can separately consider the spin and charge imbalances in steady states. As an application of the Boltzmann equations, we treat the spin imbalance in a quasi-one-dimensional superconducting wire weakly coupled with a ferromagnetic electrode through a tunnel junction. The spin imbalance induces a shift $\delta\mu$ ($-\delta\mu$) of the chemical potential for up-spin (down-spin) quasiparticles. We discuss spatial decay of $\delta\mu$ due to spin-orbit impurity scattering. At high temperatures, the decay of $\delta\mu(x)$ obeys the exponential law $\mathrm{e}^{-|x-x_0|/\lambda_{\rm s}}$, where $\lambda_{\rm s}$ is the spin-diffusion length and $|x-x_0|$ represents the distance from the injection point $x_0$. This is in agreement with the previous result.[15] However, at low temperatures, we observe deviations from the exponential law near the injection point.

In the next section, we consider a superconductor coupled with a ferromagnetic metal and introduce the quasiclassical Green's functions for the superconductor in the Keldysh formalism. The kinetic equations for them are derived in the presence of spin-orbit impurity scattering. In §3, we introduce four distribution functions to describe both the spin and charge imbalances, and derive a set of Boltzmann equations. We clarify the meaning of each distribution function. In §4, we analyze the spin-imbalance relaxation in a thin superconducting

wire based on the resulting Boltzmann equations. Section 5 is devoted to a short summary. We set $\hbar = k_{\mathrm{B}} = 1$ throughout this paper.

## 2. Kinetic Equations for Green's Functions

We consider a superconductor coupled with a ferromagnetic metal through a point-like tunnel junction. As a model for the superconductor, we adopt an electron gas interacting with phonons. Phonons are assumed to be described by the Debye model with the sound velocity $c_0$. We assume that electrons experience normal impurity scattering and spin-orbit impurity scattering. Let $\psi_\sigma(x)$ with $x \equiv (\boldsymbol{r}, t)$ be the electron field operator in the superconductor. We introduce $\Psi(x) = {}^t(\psi_\uparrow(x), \psi_\downarrow^\dagger(x))$ and define the following Green's functions[16]

$$\hat{G}^K(x,x') = -\mathrm{i}\hat{\tau}_z \left\langle \left[\Psi(x), \Psi^\dagger(x')\right]_- \right\rangle, \tag{1}$$

$$\hat{G}^R(x,x') = -\mathrm{i}\hat{\tau}_z \Theta(t-t') \left\langle \left[\Psi(x), \Psi^\dagger(x')\right]_+ \right\rangle, \tag{2}$$

$$\hat{G}^A(x,x') = +\mathrm{i}\hat{\tau}_z \Theta(t'-t) \left\langle \left[\Psi(x), \Psi^\dagger(x')\right]_+ \right\rangle, \tag{3}$$

where $\Theta(t)$ is Heavisid's step function. Here and hereafter, $\hat{\tau}_i$ $(i = x, y, z)$ represents the Pauli matrix. We use the Keldysh representation

$$\underline{G}(x,x') = \begin{pmatrix} \hat{G}^R(x,x') & \hat{G}^K(x,x') \\ 0 & \hat{G}^A(x,x') \end{pmatrix}, \tag{4}$$

and define its Fourier transform as

$$\underline{G}(\boldsymbol{r}, \boldsymbol{p}, t, t') \equiv \int \mathrm{d}^3 s \, \mathrm{e}^{-\mathrm{i}ps} \underline{G}\left(\boldsymbol{r} + \frac{\boldsymbol{s}}{2}, t, \boldsymbol{r} - \frac{\boldsymbol{s}}{2}, t'\right). \tag{5}$$

Integrating this over $\xi \equiv \boldsymbol{p}^2/(2m) - \mu$, we obtain the quasiclassical Green's function[17]

$$\underline{G}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') = \frac{\mathrm{i}}{\pi} \int \mathrm{d}\xi \, \underline{G}(\boldsymbol{r}, \boldsymbol{p}, t, t'), \tag{6}$$

where $\hat{\boldsymbol{p}} = \boldsymbol{p}/|\boldsymbol{p}|$. It has been shown that the quasiclassical Green's function obeys[18,19]

$$\begin{aligned} &v_{\mathrm{F}} \hat{\boldsymbol{p}} \cdot \nabla \underline{G}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') + \underline{\tau}_z \partial_t \underline{G}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') + \partial_{t'} \underline{G}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') \underline{\tau}_z \\ &\quad + \mathrm{i} \int_{-\infty}^{\infty} \mathrm{d}t_1 \left( \underline{\Sigma}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t_1) \underline{G}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t_1, t') - \underline{G}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t_1) \underline{\Sigma}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t_1, t') \right) = 0, \end{aligned} \tag{7}$$

where $\underline{\tau}_i \equiv \mathrm{diag}(\hat{\tau}_i, \hat{\tau}_i)$ and $\underline{\Sigma}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t')$ represents the self-energy part. The self-energy part is decomposed into

$$\underline{\Sigma}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') = \underline{\Sigma}_{\mathrm{imp}}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') + \underline{\Sigma}_{\mathrm{so}}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') + \underline{\Sigma}_{\mathrm{ph}}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') + \underline{\Sigma}_{\mathrm{inj}}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t'), \tag{8}$$

where $\underline{\Sigma}_{\mathrm{imp}}$, $\underline{\Sigma}_{\mathrm{so}}$ and $\underline{\Sigma}_{\mathrm{ph}}$ represent the contributions from normal impurity scattering, spin-orbit impurity scattering and electron-phonon interaction, respectively. It should be noted that $\underline{\Sigma}_{\mathrm{ph}}$ contains the pair potential. The last term $\underline{\Sigma}_{\mathrm{inj}}$ represents the spin-polarized current injection from the ferromagnetic metal. We simplify eq. (7) by following the argument by Usadel.[20] We employ an approximation

$$\underline{G}(\boldsymbol{r}, \hat{\boldsymbol{p}}, t, t') = \underline{G}(\boldsymbol{r}, t, t') + \hat{\boldsymbol{p}} \cdot \underline{G}_1(\boldsymbol{r}, t, t'), \tag{9}$$

where $\underline{G}_1(\boldsymbol{r},t,t')$ is much smaller then $\underline{G}(\boldsymbol{r},t,t')$. After some manipulations, we obtain

$$D\nabla\cdot\left(\underline{G}(\boldsymbol{r},\epsilon,t)\nabla\underline{G}(\boldsymbol{r},\epsilon,t)\right)+\mathrm{i}\epsilon\left[\underline{\tau}_z,\underline{G}(\boldsymbol{r},\epsilon,t)\right]_- \\ -\frac{1}{2}\partial_t\left[\underline{\tau}_z,\underline{G}(\boldsymbol{r},\epsilon,t)\right]_+-\mathrm{i}\left[\underline{\Sigma}(\boldsymbol{r},\epsilon,t),\underline{G}(\boldsymbol{r},\epsilon,t)\right]_-=0, \tag{10}$$

where

$$\underline{G}(\boldsymbol{r},\epsilon,t)=\int \mathrm{d}s\ \mathrm{e}^{\mathrm{i}\epsilon s}\underline{G}\left(\boldsymbol{r},t+\frac{s}{2},t-\frac{s}{2}\right). \tag{11}$$

In the following, we restrict our consideration to steady states, and neglect the $t$-dependence of all the functions.

We evaluate the elements of the self-energy part. We first consider $\underline{\Sigma}_{\mathrm{imp}}$ and $\underline{\Sigma}_{\mathrm{so}}$. The Hamiltonian $H_{\mathrm{imp}}$ for normal impurity scattering is written as

$$H_{\mathrm{imp}}=\sum_{\sigma}\int \mathrm{d}^3r\ \psi^\dagger_\sigma(\boldsymbol{r})V_{\mathrm{imp}}(\boldsymbol{r})\psi_\sigma(\boldsymbol{r}). \tag{12}$$

We assume for simplicity that the impurity potential is given by

$$V_{\mathrm{imp}}(\boldsymbol{r})=u_{\mathrm{imp}}\sum_j\delta(\boldsymbol{r}-\boldsymbol{R}_j), \tag{13}$$

where $\boldsymbol{R}_j$ indicates the position of the $j$th impurity. The Hamiltonian $H_{\mathrm{so}}$ for spin-orbit impurity scattering is written as

$$H_{\mathrm{so}}=\sum_{\sigma,\sigma'}\int \mathrm{d}^3r\ \psi^\dagger_\sigma(\boldsymbol{r})U^{\sigma,\sigma'}_{\mathrm{so}}(\boldsymbol{r})\psi_{\sigma'}(\boldsymbol{r}). \tag{14}$$

The spin-dependent potential is

$$U^{\sigma,\sigma'}_{\mathrm{so}}(\boldsymbol{r})=-\mathrm{i}(\vec{\hat{\tau}})_{\sigma,\sigma'}\cdot\left(\nabla V_{\mathrm{so}}(\boldsymbol{r})\times\nabla\right), \tag{15}$$

where $V_{\mathrm{so}}(\boldsymbol{r})$ is given by eq. (13) with the replacement of $u_{\mathrm{imp}}\to u_{\mathrm{so}}/k_{\mathrm{F}}^2$. In this case, $\underline{\Sigma}^{\mathrm{imp}}$ and $\underline{\Sigma}^{\mathrm{so}}$ are given by

$$\underline{\Sigma}_{\mathrm{imp}}(\boldsymbol{r},\epsilon)=-\frac{\mathrm{i}}{2\tau_{\mathrm{imp}}}\underline{G}(\boldsymbol{r},\epsilon), \tag{16}$$

$$\underline{\Sigma}_{\mathrm{so}}(\boldsymbol{r},\epsilon)=-\frac{\mathrm{i}}{6\tau_{\mathrm{so}}}\underline{G}(\boldsymbol{r},\epsilon)+\frac{\mathrm{i}}{3\tau_{\mathrm{so}}}\underline{\tau}_y\underline{G}'(\boldsymbol{r},\epsilon)\underline{\tau}_y. \tag{17}$$

The relaxation times are given by

$$\frac{1}{\tau_{\mathrm{imp}}}=2\pi n_{\mathrm{imp}}N_{\mathrm{S}}(0)|u_{\mathrm{imp}}|^2, \tag{18}$$

$$\frac{1}{\tau_{\mathrm{so}}}=2\pi n_{\mathrm{imp}}N_{\mathrm{S}}(0)|u_{\mathrm{so}}|^2\sum_{i=x,y,z}\overline{(\hat{\boldsymbol{p}}\times\hat{\boldsymbol{p}}')_i^2}, \tag{19}$$

where $n_{\mathrm{imp}}$ and $N_{\mathrm{S}}(0)$ represent the impurity concentration and the density of states in the normal state, respectively. Note that the second term of $\underline{\Sigma}^{\mathrm{so}}$ with $\underline{G}'$ represents spin-flip processes. The Green's function $\underline{G}'(\boldsymbol{r},\epsilon)$ is the Fourier transform of $\underline{G}'(x,x')$, where $\underline{G}'(x,x')$

is defined by eqs. (1)-(4) with the replacement $\Psi(x) \to \Psi'(x) = {}^t(\psi^\dagger_\uparrow(x), \psi_\downarrow(x))$. We can show that its elements $\hat{G}'^X(x, x')$ $(X = R, A, K)$ satisfy

$$\hat{G}'^X(x, x') = -\hat{\tau}_z \, {}^t[\hat{G}^{\bar{X}}(x', x)]\hat{\tau}_z, \tag{20}$$

where $\bar{R} = A$, $\bar{A} = R$ and $\bar{K} = K$. These relation will be used later. We obtain

$$\underline{\Sigma}_{\rm imp}(\boldsymbol{r}, \epsilon) + \underline{\Sigma}_{\rm so}(\boldsymbol{r}, \epsilon) = -\frac{\rm i}{2\tau}\underline{G}(\boldsymbol{r}, \epsilon) - \frac{\rm i}{3\tau_{\rm so}}\left(-\underline{\tau}_y \underline{G}'(\boldsymbol{r}, \epsilon)\underline{\tau}_y - \underline{G}(\boldsymbol{r}, \epsilon)\right) \tag{21}$$

with $\tau^{-1} \equiv \tau_{\rm imp}^{-1} + \tau_{\rm so}^{-1}$.

We turn to $\underline{\Sigma}_{\rm inj}$. Let us consider the case where a ferromagnetic metal is coupled to the superconductor through a point-like tunnel junction at $\boldsymbol{r}_0$. The corresponding Hamiltonian $H_{\rm inj}$ is given by

$$H_{\rm inj} = \sum_\sigma \int {\rm d}^3 r {\rm d}^3 r' \left(\mathcal{T}(\boldsymbol{r}, \boldsymbol{r}')\psi^\dagger_\sigma(\boldsymbol{r})\phi_\sigma(\boldsymbol{r}'){\rm e}^{-{\rm i}eVt} + {\rm h.c.}\right), \tag{22}$$

where $\phi_\sigma$ and $V$ represent the electron field operator in the ferromagnetic metal and an applied bias voltage, respectively, and we assume that $\mathcal{T}(\boldsymbol{r}, \boldsymbol{r}') = \mathcal{T}\delta(\boldsymbol{r} - \boldsymbol{r}_0)\delta(\boldsymbol{r}' - \boldsymbol{r}'_0)$. With this model, we obtain

$$\hat{\Sigma}^X_{\rm inj}(\boldsymbol{r}, \epsilon) = -{\rm i}|\mathcal{T}|^2\delta(\boldsymbol{r} - \boldsymbol{r}_0)\hat{\sigma}^X_{\rm inj}(\epsilon) \tag{23}$$

with

$$\hat{\sigma}^{R,A}_{\rm inj}(\epsilon) = \pm\begin{pmatrix} N_{{\rm F}\uparrow}(0) & 0 \\ 0 & -N_{{\rm F}\downarrow}(0) \end{pmatrix}, \tag{24}$$

$$\hat{\sigma}^K_{\rm inj}(\epsilon) = 2\begin{pmatrix} \tanh\left(\frac{\epsilon - eV}{2T}\right)N_{{\rm F}\uparrow}(0) & 0 \\ 0 & -\tanh\left(\frac{\epsilon + eV}{2T}\right)N_{{\rm F}\downarrow}(0) \end{pmatrix}, \tag{25}$$

where $N_{{\rm F}\uparrow}(0)$ $(N_{{\rm F}\downarrow}(0))$ represents the density of states for up-spin (down-spin) electrons in the ferromagnetic metal.

The phonon self-energy part has been presented in ref 18. Note that $\hat{\Sigma}^R_{\rm ph}$ and $\hat{\Sigma}^A_{\rm ph}$ contain the pair potential, so we separate it out as

$$\hat{\Sigma}^{R,A}_{\rm ph}(\boldsymbol{r}, \epsilon) \to \hat{\Sigma}^{R,A}_{\rm ph}(\boldsymbol{r}, \epsilon) - {\rm i}\hat{\tau}_\chi(\boldsymbol{r})\Delta(\boldsymbol{r}) \tag{26}$$

with

$$\hat{\tau}_\chi(\boldsymbol{r}) \equiv \cos\chi(\boldsymbol{r})\hat{\tau}_y + \sin\chi(\boldsymbol{r})\hat{\tau}_x, \tag{27}$$

where $\chi(\boldsymbol{r})$ and $\Delta(\boldsymbol{r})$ represent the phase and amplitude of the pair potential, respectively. Assuming that phonons are in thermal equilibrium, we obtain

$$\hat{\Sigma}^{R,A}_{\rm ph}(\boldsymbol{r}, \epsilon) = -{\rm i}\int {\rm d}\epsilon' \sigma_{\rm ph}(\epsilon, \epsilon')\left\{\coth\left(\frac{\epsilon' - \epsilon}{2T}\right)\hat{G}^{R,A}(\boldsymbol{r}, \epsilon') \mp \frac{1}{2}\hat{G}^K(\boldsymbol{r}, \epsilon')\right\}, \tag{28}$$

$$\hat{\Sigma}^K_{\rm ph}(\boldsymbol{r}, \epsilon) = -{\rm i}\int {\rm d}\epsilon' \sigma_{\rm ph}(\epsilon, \epsilon')\left\{\coth\left(\frac{\epsilon' - \epsilon}{2T}\right)\hat{G}^K(\boldsymbol{r}, \epsilon') - \left(\hat{G}^R(\boldsymbol{r}, \epsilon') - \hat{G}^A(\boldsymbol{r}, \epsilon')\right)\right\}, \tag{29}$$

where

$$\sigma_{\mathrm{ph}}(\epsilon,\epsilon') = \frac{N_{\mathrm{S}}(0)g^2}{8} \cdot \frac{\pi}{(c_0 k_{\mathrm{F}})^2}(\epsilon'-\epsilon)^2 \mathrm{sign}(\epsilon'-\epsilon). \tag{30}$$

Here, $c_0$ and $g$ represent the sound velocity and the coupling constant for the electron-phonon interaction, respectively.

Using eq. (10) and the elements of the self-energy part, we obtain the kinetic equations for the Green's functions,

$$D\nabla \cdot \left(\hat{G}^{R,A}\nabla\hat{G}^{R,A}\right) + \mathrm{i}\epsilon\big[\hat{\tau}_z, \hat{G}^{R,A}\big]_- - \Delta\big[\hat{\tau}_\chi, \hat{G}^{R,A}\big]_- + \hat{\Gamma}^{R,A} - \hat{I}^{R,A} - \hat{P}^{R,A} = 0, \tag{31}$$

$$D\nabla \cdot \left(\hat{G}^{R}\nabla\hat{G}^{K} + \hat{G}^{K}\nabla\hat{G}^{A}\right) + \mathrm{i}\epsilon\big[\hat{\tau}_z, \hat{G}^{K}\big]_- - \Delta\big[\hat{\tau}_\chi, \hat{G}^{K}\big]_- + \hat{\Gamma}^{K} - \hat{I}^{K} - \hat{P}^{K} = 0, \tag{32}$$

where

$$\hat{\Gamma}^{R,A} = \frac{1}{3\tau_{\mathrm{so}}}\big(\hat{\tau}_y\hat{G}'^{R,A}\hat{\tau}_y\hat{G}^{R,A} - \hat{G}^{R,A}\hat{\tau}_y\hat{G}'^{R,A}\hat{\tau}_y\big), \tag{33}$$

$$\hat{\Gamma}^{K} = \frac{1}{3\tau_{\mathrm{so}}}\big(\hat{\tau}_y\hat{G}'^{R}\hat{\tau}_y\hat{G}^{K} + \hat{\tau}_y\hat{G}'^{K}\hat{\tau}_y\hat{G}^{A} - \hat{G}^{R}\hat{\tau}_y\hat{G}'^{K}\hat{\tau}_y - \hat{G}^{K}\hat{\tau}_y\hat{G}'^{A}\hat{\tau}_y\big), \tag{34}$$

$$\hat{I}^{R,A} = \mathrm{i}\big(\hat{\Sigma}^{R,A}_{\mathrm{ph}}\hat{G}^{R,A} - \hat{G}^{R,A}\hat{\Sigma}^{R,A}_{\mathrm{ph}}\big), \tag{35}$$

$$\hat{I}^{K} = \mathrm{i}\big(\hat{\Sigma}^{R}_{\mathrm{ph}}\hat{G}^{K} + \hat{\Sigma}^{K}_{\mathrm{ph}}\hat{G}^{A} - \hat{G}^{R}\hat{\Sigma}^{K}_{\mathrm{ph}} - \hat{G}^{K}\hat{\Sigma}^{A}_{\mathrm{ph}}\big), \tag{36}$$

and $\hat{P}^X$ is obtained by the replacement of $\hat{\Sigma}^X_{\mathrm{ph}} \to \hat{\Sigma}^X_{\mathrm{inj}}$ in $\hat{I}^X$. The arguments $\boldsymbol{r}$ and $\epsilon$ are suppressed in the above equations. Here, $\hat{\Gamma}^X$ represent the influence of spin-flip processes due to spin-orbit impurity scattering, and $\hat{I}^X$ and $\hat{P}^X$ describe inelastic phonon scattering and a spin-polarized current injection, respectively. If we neglect $\hat{\Gamma}^X$ and set $N_{\mathrm{F}\uparrow}(0) = N_{\mathrm{F}\downarrow}(0)$ in $\hat{P}^X$, our argument is reduced to the previous one presented by Schmid and Schön.[7] Quasiparticle distribution functions are contained in $\hat{G}^K$ and $\hat{G}'^K$.

## 3. Boltzmann Equations

Based on the kinetic equations presented in the previous section, we start to derive a set of linearized Boltzmann equations which describes a nonequilibrium quasiparticle distribution in superconductors. In terms of the spectral functions $N_1$, $N_2$, $R_1$ and $R_2$, we approximately express $\hat{G}^R$ and $\hat{G}^A$ as[7]

$$\hat{G}^{R,A}(\boldsymbol{r},\epsilon) = \big(\pm N_1(\boldsymbol{r},\epsilon) + \mathrm{i}R_1(\boldsymbol{r},\epsilon)\big)\hat{\tau}_z + \big(N_2(\boldsymbol{r},\epsilon) \pm \mathrm{i}R_2(\boldsymbol{r},\epsilon)\big)\hat{\tau}_\chi(\boldsymbol{r}). \tag{37}$$

The spectral functions satisfy

$$N_{1,2}(\boldsymbol{r},\epsilon) = N_{1,2}(\boldsymbol{r},-\epsilon), \tag{38}$$

$$R_{1,2}(\boldsymbol{r},\epsilon) = -R_{1,2}(\boldsymbol{r},-\epsilon). \tag{39}$$

We rewrite $\hat{\tau}_y\hat{G}'^X(\boldsymbol{r},\epsilon)\hat{\tau}_y$ in terms of $\hat{G}^X(\boldsymbol{r},\epsilon)$. From eq. (20), we obtain

$$\hat{G}'^X(\boldsymbol{r},\epsilon) = -\hat{\tau}_z\, {}^t[\hat{G}^{\bar{X}}(\boldsymbol{r},-\epsilon)]\, \hat{\tau}_z. \tag{40}$$

Combining eqs. (37) and (40) with eqs. (38) and (39), we obtain $\hat{\tau}_y\hat{G}'^R(\boldsymbol{r},\epsilon)\hat{\tau}_y = -\hat{G}^R(\boldsymbol{r},\epsilon)$ and $\hat{\tau}_y\hat{G}'^A(\boldsymbol{r},\epsilon)\hat{\tau}_y = -\hat{G}^A(\boldsymbol{r},\epsilon)$. Note that $\hat{\Gamma}^{R,A} = 0$ if these results are substituted into eq. (33). This indicates that spin-flip processes due to spin-orbit impurity scattering do not influence on the spectral properties.[21] Only quasiparticle distribution functions contained in $\hat{G}^K$ are affected by spin-flip processes. We adopt the following expression[19]

$$\hat{G}^K(\boldsymbol{r},\epsilon) = \hat{G}^R(\boldsymbol{r},\epsilon)\hat{h}(\boldsymbol{r},\epsilon) - \hat{h}(\boldsymbol{r},\epsilon)\hat{G}^A(\boldsymbol{r},\epsilon) \tag{41}$$

with

$$\hat{h}(\boldsymbol{r},\epsilon) = h_1(\boldsymbol{r},\epsilon) + h_2(\boldsymbol{r},\epsilon)\hat{\tau}_z. \tag{42}$$

A nonequilibrium quasiparticle distribution is described by $h_1$ and $h_2$. From eq. (40), we obtain

$$\hat{\tau}_y\hat{G}'^K(\boldsymbol{r},\epsilon)\hat{\tau}_y = \hat{G}^R(\boldsymbol{r},\epsilon)\hat{\tau}_y\hat{h}(\boldsymbol{r},-\epsilon)\hat{\tau}_y - \hat{\tau}_y\hat{h}(\boldsymbol{r},-\epsilon)\hat{\tau}_y\hat{G}^A(\boldsymbol{r},\epsilon). \tag{43}$$

By using eqs. (41) and (43), we simplify the expression of $\hat{\Gamma}^K$ as

$$\hat{\Gamma}^K(\boldsymbol{r},\epsilon) = \frac{2}{3\tau_{\mathrm{so}}}\left(\hat{G}^R(\boldsymbol{r},\epsilon)\delta\hat{h}(\boldsymbol{r},\epsilon)\hat{G}^A(\boldsymbol{r},\epsilon) - \delta\hat{h}(\boldsymbol{r},\epsilon)\right) \tag{44}$$

with

$$\delta\hat{h}(\boldsymbol{r},\epsilon) = \left(h_1(\boldsymbol{r},\epsilon) + h_1(\boldsymbol{r},-\epsilon)\right) + \left(h_2(\boldsymbol{r},\epsilon) - h_2(\boldsymbol{r},-\epsilon)\right)\hat{\tau}_z. \tag{45}$$

At equilibrium, $h_{1,2}(\boldsymbol{r},\epsilon)$ is reduced to $h_1 = \tanh(\epsilon/(2T))$ and $h_2 = 0$. It is convenient to set

$$h_1(\boldsymbol{r},\epsilon) = \tanh\left(\frac{\epsilon}{2T}\right) - 2f_{\mathrm{L}}(\boldsymbol{r},\epsilon), \tag{46}$$

$$h_2(\boldsymbol{r},\epsilon) = -2f_{\mathrm{T}}(\boldsymbol{r},\epsilon). \tag{47}$$

The quasiparticle distribution functions are written as[7]

$$f_{\uparrow}(\boldsymbol{r},\epsilon) = f_{\mathrm{FD}}(\epsilon,\mu) + f_{\mathrm{L}}(\boldsymbol{r},\epsilon) + f_{\mathrm{T}}(\boldsymbol{r},\epsilon), \tag{48}$$

$$f_{\downarrow}(\boldsymbol{r},\epsilon) = f_{\mathrm{FD}}(\epsilon,\mu) - f_{\mathrm{L}}(\boldsymbol{r},-\epsilon) + f_{\mathrm{T}}(\boldsymbol{r},-\epsilon), \tag{49}$$

where $f_{\mathrm{FD}}(\epsilon,\mu)$ is the Fermi-Dirac distribution function. It should be emphasized that $f_{\mathrm{L}}(\boldsymbol{r},\epsilon) = -f_{\mathrm{L}}(\boldsymbol{r},-\epsilon)$ and $f_{\mathrm{T}}(\boldsymbol{r},\epsilon) = f_{\mathrm{T}}(\boldsymbol{r},-\epsilon)$ are implicitly assumed in ref. 7. These relations straightforwardly result in $f_{\uparrow}(\boldsymbol{r},\epsilon) = f_{\downarrow}(\boldsymbol{r},\epsilon)$. Thus, the framework by Schmid and Schön is restricted to spin-independent phenomena. We do not accept the symmetry relations to enable us to consider the spin imbalance.

We obtain Boltzmann equations based on eqs. (31) and (44). Substituting eqs. (41) and (43) into eq. (32), we derive equations for $f_{\mathrm{L}}(\boldsymbol{r},\epsilon)$ and $f_{\mathrm{T}}(\boldsymbol{r},\epsilon)$ with the help of eq. (31).

Details of the derivation are described in ref. 16. We obtain

$$D \cdot \nabla\big(N_1^2(\boldsymbol{r},\epsilon) - R_2^2(\boldsymbol{r},\epsilon)\big)\nabla f_{\mathrm{L}}(\boldsymbol{r},\epsilon) - \frac{2}{3\tau_{\mathrm{so}}}\big(N_1^2(\boldsymbol{r},\epsilon) - R_2^2(\boldsymbol{r},\epsilon)\big)\big(f_{\mathrm{L}}(\boldsymbol{r},\epsilon) + f_{\mathrm{L}}(\boldsymbol{r},-\epsilon)\big)$$
$$+ I_{\mathrm{L}}\big(\boldsymbol{r},\epsilon,\{f_{\mathrm{L}}\}\big) + P_{\mathrm{L}}(\boldsymbol{r},\epsilon) = 0, \tag{50}$$

$$D \cdot \nabla\big(N_1^2(\boldsymbol{r},\epsilon) + N_2^2(\boldsymbol{r},\epsilon)\big)\nabla f_{\mathrm{T}}(\boldsymbol{r},\epsilon) - \frac{2}{3\tau_{\mathrm{so}}}\big(N_1^2(\boldsymbol{r},\epsilon) + N_2^2(\boldsymbol{r},\epsilon)\big)\big(f_{\mathrm{T}}(\boldsymbol{r},\epsilon) - f_{\mathrm{T}}(\boldsymbol{r},-\epsilon)\big)$$
$$- \frac{1}{\tau_{\mathrm{conv}}(\epsilon)} f_{\mathrm{T}}(\boldsymbol{r},\epsilon) + I_{\mathrm{T}}\big(\boldsymbol{r},\epsilon,\{f_{\mathrm{T}}\}\big) + P_{\mathrm{T}}(\boldsymbol{r},\epsilon) = 0, \tag{51}$$

where $\tau_{\mathrm{conv}}(\epsilon)$ represents the conversion time for charge imbalance.[22] The collision integrals $I_{\mathrm{L}}$ and $I_{\mathrm{T}}$ due to inelastic phonon scattering are expressed as

$$I_{\mathrm{L,T}}\big(\boldsymbol{r},\epsilon,\{f\}\big) = -2\int \mathrm{d}\epsilon' \sigma_{\mathrm{ph}}(\epsilon,\epsilon') M_{\mathrm{L,T}}(\boldsymbol{r},\epsilon,\epsilon')$$
$$\times \frac{\cosh^2\big(\frac{\epsilon}{2T}\big) f(\boldsymbol{r},\epsilon) - \cosh^2\big(\frac{\epsilon'}{2T}\big) f(\boldsymbol{r},\epsilon')}{\sinh\big(\frac{\epsilon'-\epsilon}{2T}\big)\cosh\big(\frac{\epsilon}{2T}\big)\cosh\big(\frac{\epsilon'}{2T}\big)}, \tag{52}$$

where

$$M_{\mathrm{L}}(\boldsymbol{r},\epsilon,\epsilon') = N_1(\boldsymbol{r},\epsilon)N_1(\boldsymbol{r},\epsilon') - R_2(\boldsymbol{r},\epsilon)R_2(\boldsymbol{r},\epsilon'), \tag{53}$$

$$M_{\mathrm{T}}(\boldsymbol{r},\epsilon,\epsilon') = N_1(\boldsymbol{r},\epsilon)N_1(\boldsymbol{r},\epsilon') + N_2(\boldsymbol{r},\epsilon)N_2(\boldsymbol{r},\epsilon'). \tag{54}$$

The injection terms $P_{\mathrm{L}}$ and $P_{\mathrm{T}}$ are given by

$$P_{\mathrm{L}}(\boldsymbol{r},\epsilon) = \frac{\pi}{2}|\mathcal{T}|^2\delta(\boldsymbol{r}-\boldsymbol{r}_0)N_1(\boldsymbol{r},\epsilon)\Big\{N_{\mathrm{F}\uparrow}(0)\Big(\tanh\big(\frac{\epsilon}{2T}\big) - \tanh\big(\frac{\epsilon - eV}{2T}\big)\Big)$$
$$+ N_{\mathrm{F}\downarrow}(0)\Big(\tanh\big(\frac{\epsilon}{2T}\big) - \tanh\big(\frac{\epsilon + eV}{2T}\big)\Big)\Big\}, \tag{55}$$

$$P_{\mathrm{T}}(\boldsymbol{r},\epsilon) = \frac{\pi}{2}|\mathcal{T}|^2\delta(\boldsymbol{r}-\boldsymbol{r}_0)N_1(\boldsymbol{r},\epsilon)\Big\{N_{\mathrm{F}\uparrow}(0)\Big(\tanh\big(\frac{\epsilon}{2T}\big) - \tanh\big(\frac{\epsilon - eV}{2T}\big)\Big)$$
$$- N_{\mathrm{F}\downarrow}(0)\Big(\tanh\big(\frac{\epsilon}{2T}\big) - \tanh\big(\frac{\epsilon + eV}{2T}\big)\Big)\Big\}. \tag{56}$$

To make the equations much simpler, we introduce the four distribution functions,

$$f_{\mathrm{L}+}(\boldsymbol{r},\epsilon) = \frac{1}{2}\left(f_{\mathrm{L}}(\boldsymbol{r},\epsilon) + f_{\mathrm{L}}(\boldsymbol{r},-\epsilon)\right), \tag{57}$$

$$f_{\mathrm{L}-}(\boldsymbol{r},\epsilon) = \frac{1}{2}\left(f_{\mathrm{L}}(\boldsymbol{r},\epsilon) - f_{\mathrm{L}}(\boldsymbol{r},-\epsilon)\right), \tag{58}$$

$$f_{\mathrm{T}+}(\boldsymbol{r},\epsilon) = \frac{1}{2}\left(f_{\mathrm{T}}(\boldsymbol{r},\epsilon) + f_{\mathrm{T}}(\boldsymbol{r},-\epsilon)\right), \tag{59}$$

$$f_{\mathrm{T}-}(\boldsymbol{r},\epsilon) = \frac{1}{2}\left(f_{\mathrm{T}}(\boldsymbol{r},\epsilon) - f_{\mathrm{T}}(\boldsymbol{r},-\epsilon)\right). \tag{60}$$

We observe that they satisfy

$$f_{\mathrm{L,T}+}(\boldsymbol{r},-\epsilon) = f_{\mathrm{L,T}+}(\boldsymbol{r},\epsilon), \tag{61}$$

$$f_{\mathrm{L,T}-}(\boldsymbol{r},-\epsilon) = -f_{\mathrm{L,T}-}(\boldsymbol{r},\epsilon). \tag{62}$$

Noting eqs. (30), (38) and (39), we obtain a set of Boltzmann equations for $f_{\mathrm{L}\pm}$ and $f_{\mathrm{T}\pm}$ as

$$D\cdot\nabla\big(N_1^2(\boldsymbol{r},\epsilon)-R_2^2(\boldsymbol{r},\epsilon)\big)\nabla f_{\mathrm{L}+}(\boldsymbol{r},\epsilon) - \frac{1}{\tau_{\mathrm{sf}}}\big(N_1^2(\boldsymbol{r},\epsilon)-R_2^2(\boldsymbol{r},\epsilon)\big)f_{\mathrm{L}+}(\boldsymbol{r},\epsilon)+I_{\mathrm{L}}\big(\boldsymbol{r},\epsilon,\{f_{\mathrm{L}+}\}\big)+P_{\mathrm{L}+}(\boldsymbol{r},\epsilon)=0, \tag{63}$$

$$D\cdot\nabla\big(N_1^2(\boldsymbol{r},\epsilon)-R_2^2(\boldsymbol{r},\epsilon)\big)\nabla f_{\mathrm{L}-}(\boldsymbol{r},\epsilon)+I_{\mathrm{L}}\big(\boldsymbol{r},\epsilon,\{f_{\mathrm{L}-}\}\big)+P_{\mathrm{L}-}(\boldsymbol{r},\epsilon)=0, \tag{64}$$

$$D\cdot\nabla\big(N_1^2(\boldsymbol{r},\epsilon)+N_2^2(\boldsymbol{r},\epsilon)\big)\nabla f_{\mathrm{T}+}(\boldsymbol{r},\epsilon)-\frac{1}{\tau_{\mathrm{conv}}(\epsilon)}f_{\mathrm{T}+}(\boldsymbol{r},\epsilon)+I_{\mathrm{T}}\big(\boldsymbol{r},\epsilon,\{f_{\mathrm{T}+}\}\big)+P_{\mathrm{T}+}(\boldsymbol{r},\epsilon)=0, \tag{65}$$

$$D\cdot\nabla\big(N_1^2(\boldsymbol{r},\epsilon)+N_2^2(\boldsymbol{r},\epsilon)\big)\nabla f_{\mathrm{T}-}(\boldsymbol{r},\epsilon)-\frac{1}{\tau_{\mathrm{conv}}(\epsilon)}f_{\mathrm{T}-}(\boldsymbol{r},\epsilon) - \frac{1}{\tau_{\mathrm{sf}}}\big(N_1^2(\boldsymbol{r},\epsilon)+N_2^2(\boldsymbol{r},\epsilon)\big)f_{\mathrm{T}-}(\boldsymbol{r},\epsilon)+I_{\mathrm{T}}\big(\boldsymbol{r},\epsilon,\{f_{\mathrm{T}-}\}\big)+P_{\mathrm{T}-}(\boldsymbol{r},\epsilon)=0, \tag{66}$$

where $\tau_{\mathrm{sf}}^{-1}=(4/3)\tau_{\mathrm{so}}^{-1}$ and

$$P_{\mathrm{L}\pm}(\boldsymbol{r},\epsilon)=\frac{1}{2}\left(P_{\mathrm{L}}(\boldsymbol{r},\epsilon)\pm P_{\mathrm{L}}(\boldsymbol{r},-\epsilon)\right), \tag{67}$$

$$P_{\mathrm{T}\pm}(\boldsymbol{r},\epsilon)=\frac{1}{2}\left(P_{\mathrm{T}}(\boldsymbol{r},\epsilon)\pm P_{\mathrm{T}}(\boldsymbol{r},-\epsilon)\right). \tag{68}$$

Using the expression of the tunnel resistance,

$$R_t^{-1}=2\pi e^2N_{\mathrm{S}}(0)\big(N_{\mathrm{F}\uparrow}(0)+N_{\mathrm{F}\downarrow}(0)\big)|\mathcal{T}|^2, \tag{69}$$

we can rewrite the injection terms as

$$P_{\mathrm{L}+}(\boldsymbol{r},\epsilon)=\frac{P_{\mathrm{s}}N_1(\boldsymbol{r},\epsilon)}{8e^2N_{\mathrm{S}}(0)R_t}\delta(\boldsymbol{r}-\boldsymbol{r}_0)\Big(\tanh\big(\frac{\epsilon+eV}{2T}\big)-\tanh\big(\frac{\epsilon-eV}{2T}\big)\Big), \tag{70}$$

$$P_{\mathrm{L}-}(\boldsymbol{r},\epsilon)=\frac{N_1(\boldsymbol{r},\epsilon)}{8e^2N_{\mathrm{S}}(0)R_t}\delta(\boldsymbol{r}-\boldsymbol{r}_0)\Big(2\tanh\big(\frac{\epsilon}{2T}\big)-\tanh\big(\frac{\epsilon+eV}{2T}\big)-\tanh\big(\frac{\epsilon-eV}{2T}\big)\Big), \tag{71}$$

$$P_{\mathrm{T}+}(\boldsymbol{r},\epsilon)=\frac{N_1(\boldsymbol{r},\epsilon)}{8e^2N_{\mathrm{S}}(0)R_t}\delta(\boldsymbol{r}-\boldsymbol{r}_0)\Big(\tanh\big(\frac{\epsilon+eV}{2T}\big)-\tanh\big(\frac{\epsilon-eV}{2T}\big)\Big), \tag{72}$$

$$P_{\mathrm{T}-}(\boldsymbol{r},\epsilon)=\frac{P_{\mathrm{s}}N_1(\boldsymbol{r},\epsilon)}{8e^2N_{\mathrm{S}}(0)R_t}\delta(\boldsymbol{r}-\boldsymbol{r}_0)\Big(2\tanh\big(\frac{\epsilon}{2T}\big)-\tanh\big(\frac{\epsilon+eV}{2T}\big)-\tanh\big(\frac{\epsilon-eV}{2T}\big)\Big), \tag{73}$$

where the spin polarization $P_{\mathrm{s}}$ is defined by

$$P_{\mathrm{s}}=\frac{N_{\mathrm{F}\uparrow}(0)-N_{\mathrm{F}\downarrow}(0)}{N_{\mathrm{F}\uparrow}(0)+N_{\mathrm{F}\downarrow}(0)}. \tag{74}$$

Equations (63)-(66) and eqs. (70)-(73) are the central result of this paper. We here clarify the meaning of each distribution function. The distribution functions $f_\sigma(\boldsymbol{r},\epsilon)$ are expressed as

$$f_\uparrow(\boldsymbol{r},\epsilon)=f_{\mathrm{FD}}(\epsilon,\mu)+f_{\mathrm{L}+}(\boldsymbol{r},\epsilon)+f_{\mathrm{L}-}(\boldsymbol{r},\epsilon)+f_{\mathrm{T}+}(\boldsymbol{r},\epsilon)+f_{\mathrm{T}-}(\boldsymbol{r},\epsilon), \tag{75}$$

$$f_\downarrow(\boldsymbol{r},\epsilon)=f_{\mathrm{FD}}(\epsilon,\mu)-f_{\mathrm{L}+}(\boldsymbol{r},\epsilon)+f_{\mathrm{L}-}(\boldsymbol{r},\epsilon)+f_{\mathrm{T}+}(\boldsymbol{r},\epsilon)-f_{\mathrm{T}-}(\boldsymbol{r},\epsilon). \tag{76}$$

Let $S(\boldsymbol{r})$ and $Q(\boldsymbol{r})$ be the spin and charge imbalances, respectively. Noting that $N_1(\boldsymbol{r},\epsilon)$ is the normalized local density of states in the superconductor, we can express

$$S(\boldsymbol{r}) = N_{\rm S}(0)\int_{-\infty}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\left(f_\uparrow(\boldsymbol{r},\epsilon) - f_\downarrow(\boldsymbol{r},\epsilon)\right), \tag{77}$$

$$Q(\boldsymbol{r}) = N_{\rm S}(0)\int_{-\infty}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\left(f_\uparrow(\boldsymbol{r},\epsilon) + f_\downarrow(\boldsymbol{r},\epsilon) - 2f_{\rm FD}(\epsilon,\mu)\right). \tag{78}$$

By using eqs. (75) and (76), we obtain

$$S(\boldsymbol{r}) = 4N_{\rm S}(0)\int_{0}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon) f_{\rm L+}(\boldsymbol{r},\epsilon), \tag{79}$$

$$Q(\boldsymbol{r}) = 4N_{\rm S}(0)\int_{0}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon) f_{\rm T+}(\boldsymbol{r},\epsilon). \tag{80}$$

Thus, $f_{\rm L+}$ and $f_{\rm T+}$ describe the spin and charge imbalances, respectively. Other two distribution functions are related to quasiparticle energies. Let $E_Q$ and $E_S$ be the excess quasiparticle energy and the energy imbalance between up- and down-spin quasiparticles, respectively. They are given by

$$E_Q(\boldsymbol{r}) = N_{\rm S}(0)\int_{-\infty}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\epsilon\left(f_\uparrow(\boldsymbol{r},\epsilon) + f_\downarrow(\boldsymbol{r},\epsilon) - 2f_{\rm FD}(\epsilon,\mu)\right), \tag{81}$$

$$E_S(\boldsymbol{r}) = N_{\rm S}(0)\int_{-\infty}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\epsilon\left(f_\uparrow(\boldsymbol{r},\epsilon) - f_\downarrow(\boldsymbol{r},\epsilon)\right). \tag{82}$$

By using eqs. (75) and (76), we obtain

$$E_Q(\boldsymbol{r}) = 4N_{\rm S}(0)\int_{0}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\epsilon f_{\rm L-}(\boldsymbol{r},\epsilon), \tag{83}$$

$$E_S(\boldsymbol{r}) = 4N_{\rm S}(0)\int_{0}^{\infty} {\rm d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\epsilon f_{\rm T-}(\boldsymbol{r},\epsilon). \tag{84}$$

Thus, $f_{\rm L-}$ and $f_{\rm T-}$ describe the excess quasiparticle energy and the energy imbalance between up- and down-spin quasiparticles, respectively. The two distribution functions $f_{\rm L+}$ and $f_{\rm T-}$ characterize the spin imbalance, while other two distribution functions $f_{\rm L-}$ and $f_{\rm T+}$ characterize the charge imbalance. The former two have not been discussed in literatures.

We approximately obtain the spectral functions. The presence of spin-orbit scattering does not result in any changes of the spectral properties of superconductors. According to the approximation adopted in eq. (37), we obtain

$$\Delta(\boldsymbol{r}) = N_{\rm S}(0)g^2\int_{0}^{\infty} {\rm d}\epsilon\, R_2(\boldsymbol{r},\epsilon)\left(\tanh(\frac{\epsilon}{2T}) - 2f_{\rm L-}(\boldsymbol{r},\epsilon)\right). \tag{85}$$

This indicates that the suppression of the pair potential due to a current injection is governed by $f_{\rm L-}$. In contrast, the variation of the phase $\chi$ is related to $f_{\rm T+}$ although this point is out of our scope. We assume that $\Delta$ and $\chi$ spatially vary much slower than $f_{\rm L\pm}$ and $f_{\rm T\pm}$. Thus, we approximate that $\nabla\hat{G}^{R,A} = 0$. Furthermore, we neglect the phonon self-energy in deriving $\hat{G}^{R,A}$. The local density of states vanishes for $|\epsilon| < \Delta(\boldsymbol{r})$ in this case, so we consider the energy

region of $|\epsilon| \geq \Delta(\boldsymbol{r})$ in the following. After these simplifications, we obtain

$$N_1(\boldsymbol{r},\epsilon) = \frac{|\epsilon|}{\sqrt{\epsilon^2 - \Delta^2(\boldsymbol{r})}}, \tag{86}$$

$$R_2(\boldsymbol{r},\epsilon) = \frac{\mathrm{sign}(\epsilon)\Delta(\boldsymbol{r})}{\sqrt{\epsilon^2 - \Delta^2(\boldsymbol{r})}}, \tag{87}$$

and $N_2(\boldsymbol{r},\epsilon) = 0$ for $|\epsilon| \geq \Delta(\boldsymbol{r})$.

In the presence of the spin and/or charge imbalances, the distribution functions $f_\sigma(\boldsymbol{r},\epsilon)$ deviate from the equilibrium ones. To characterize their deviations, we introduce the spin-dependent chemical potential $\mu_\sigma(\boldsymbol{r}) = \mu + \delta\mu_\sigma(\boldsymbol{r})$ for quasiparticles. To define $\delta\mu_\sigma(\boldsymbol{r})$, we assume that a fictitious electrode is weakly coupled to a superconductor at $\boldsymbol{r}$ through a point-like tunnel junction. In terms of a bias voltage $V_{\rm fic}$, the spin-dependent tunneling current $I_\sigma(\boldsymbol{r}, V_{\rm fic})$ is given by

$$I_{\uparrow,\downarrow}(\boldsymbol{r}, V_{\rm fic}) \propto \int_0^\infty \mathrm{d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\Big(\tanh\big(\frac{\epsilon + eV_{\rm fic}}{2T}\big) - \tanh\big(\frac{\epsilon - eV_{\rm fic}}{2T}\big) \\ - 4\left(\pm f_{\rm L+}(\boldsymbol{r},\epsilon) - f_{\rm T+}(\boldsymbol{r},\epsilon)\right)\Big). \tag{88}$$

The tunneling current $I_\sigma(\boldsymbol{r}, V_{\rm fic})$ vanishes if $eV_{\rm fic}$ is equal to $\delta\mu_\sigma(\boldsymbol{r})$. Thus, $\delta\mu_\sigma(\boldsymbol{r})$ satisfies the following equation

$$\int_0^\infty \mathrm{d}\epsilon\, N_1(\boldsymbol{r},\epsilon)\Big(\tanh\big(\frac{\epsilon + \delta\mu_{\uparrow,\downarrow}(\boldsymbol{r})}{2T}\big) - \tanh\big(\frac{\epsilon - \delta\mu_{\uparrow,\downarrow}(\boldsymbol{r})}{2T}\big) \\ - 4\left(\pm f_{\rm L+}(\boldsymbol{r},\epsilon) - f_{\rm T+}(\boldsymbol{r},\epsilon)\right)\Big) = 0. \tag{89}$$

We use the above equation as the definition of $\delta\mu_\sigma(\boldsymbol{r})$. We observe that $\delta\mu_\uparrow(\boldsymbol{r}) = \delta\mu_\downarrow(\boldsymbol{r})$ if the spin imbalance is absent (i.e., $f_{\rm L+}(\boldsymbol{r},\epsilon) = 0$), while $\delta\mu_\uparrow(\boldsymbol{r}) = -\delta\mu_\downarrow(\boldsymbol{r})$ in the absence of the charge imbalance (i.e., $f_{\rm T+}(\boldsymbol{r},\epsilon) = 0$).

## 4. Spin-Imbalance Relaxation

In this section, we study the behavior of spin-imbalance relaxation based on the Boltzmann equation for $f_{\rm L+}$. Let us consider a thin superconducting wire coupled with a ferromagnetic electrode through a tunnel junction. We assume that $f_{\rm L-}$ is very small everywhere in the superconductor, and set $\Delta(\boldsymbol{r}) = \Delta$. We neglect the charge imbalance for simplicity, and focus on a shift of the spin-dependent chemical potential. If the cross-sectional area $A$ of the wire is small enough, we are allowed to consider a one-dimensional problem of $f_{\rm L+}(x,\epsilon)$. As noted just below eq. (89), we observe that $\delta\mu(x) \equiv \delta\mu_\uparrow(x) = -\delta\mu_\uparrow(x)$ in this case. The Boltzmann equation for $f_{\rm L+}$ is reduced to

$$D\partial_x^2 f_{\rm L+}(x,\epsilon) - \frac{1}{\tau_{\rm sf}} f_{\rm L+}(x,\epsilon) + I_{\rm L}\big(x,\epsilon,\{f_{\rm L+}\}\big) + \tilde{P}_{\rm L+}(x,\epsilon) = 0, \tag{90}$$

where

$$I_{\rm L}\big(x,\epsilon,\{f\}\big) = -2\int {\rm d}\epsilon'\sigma_{\rm ph}(\epsilon,\epsilon')\,\big(N_1(\epsilon)N_1(\epsilon')-R_2(\epsilon)R_2(\epsilon')\big) \\ \times \frac{\cosh^2\big(\frac{\epsilon}{2T}\big)f(x,\epsilon)-\cosh^2\big(\frac{\epsilon'}{2T}\big)f(x,\epsilon')}{\sinh\big(\frac{\epsilon'-\epsilon}{2T}\big)\cosh\big(\frac{\epsilon}{2T}\big)\cosh\big(\frac{\epsilon'}{2T}\big)}, \tag{91}$$

$$\tilde{P}_{\rm L+}(x,\epsilon) = \frac{P_{\rm s}N_1(\epsilon)}{8e^2N_{\rm S}(0)R_tA}\delta(x-x_0)\Big(\tanh\big(\frac{\epsilon+eV}{2T}\big)-\tanh\big(\frac{\epsilon-eV}{2T}\big)\Big). \tag{92}$$

We define the energy relaxation time $\tau_{\rm ene}(\epsilon)$ as

$$\frac{1}{\tau_{\rm ene}(\epsilon)} = 2\int {\rm d}\epsilon'\sigma_{\rm ph}(\epsilon,\epsilon')\,\big(N_1(\epsilon)N_1(\epsilon')-R_2(\epsilon)R_2(\epsilon')\big)\,\frac{\cosh\big(\frac{\epsilon}{2T}\big)}{\sinh\big(\frac{\epsilon'-\epsilon}{2T}\big)\cosh\big(\frac{\epsilon'}{2T}\big)}, \tag{93}$$

in terms of which the first term in the collision integral $I_{\rm L}$ is rewritten as $-f_{\rm L+}(\boldsymbol{r},\epsilon)/\tau_{\rm ene}(\epsilon)$. Note that the energy of an injected quasiparticle is within $\Delta \le |\epsilon| \lesssim eV+T$. The behavior of the spin-imbalance relaxation depends on whether $\tau_{\rm sf}$ is longer or shorter than $\tau_{\rm ene}(\epsilon)$ in this energy range.

We first consider the high-temperature regime in which $eV \ll T$ and $\tau_{\rm ene}(\epsilon)$ is much shorter than $\tau_{\rm sf}$ for $\Delta \le |\epsilon| \lesssim T$. In this case, the energy dependence of $f_{\rm L+}$ is mainly determined by the collision-integral term. Except near the injection point (i.e., $x = x_0$), we can approximate $f_{\rm L+}(x,\epsilon) \propto 1/\cosh^2(\epsilon/(2T))$. Since eq. (89) is simplified to

$$\int_0^\infty {\rm d}\epsilon\; N_1(\epsilon)\Big(\frac{\delta\mu(x)}{T\cosh^2\big(\frac{\epsilon}{2T}\big)} - 4f_{\rm L+}(x,\epsilon)\Big) = 0, \tag{94}$$

we observe that

$$f_{\rm L+}(x,\epsilon) = \frac{\delta\mu(x)}{4T\cosh^2\big(\frac{\epsilon}{2T}\big)}. \tag{95}$$

We determine $\delta\mu(x)$ based on eq. (90). If we approximately set $N_1(\epsilon) = 1$ in $\tilde{P}_{\rm L+}(x,\epsilon)$, we obtain

$$\delta\mu(x) = \delta\mu_0\;{\rm e}^{-\frac{|x-x_0|}{\lambda_{\rm s}}}, \tag{96}$$

where $\lambda_{\rm s} = \sqrt{D\tau_{\rm sf}}$ is the spin-diffusion length and

$$\delta\mu_0 = \frac{2\lambda_{\rm s}P_{\rm s}eV}{8e^2N_{\rm S}(0)R_tAD}. \tag{97}$$

The approximation $N_1(\epsilon) \to 1$ results in an under-estimation of $\delta\mu_0$. It should be noted that the distribution function $f_{\uparrow,\downarrow}(x,\epsilon) \equiv f_{\rm FD}(\epsilon,\mu) \pm f_{\rm L+}(\epsilon,\mu)$ is expressed by the Fermi-Dirac distribution function with a shifted chemical potential as $f_{\uparrow,\downarrow}(x,\epsilon) \approx f_{\rm FD}(\epsilon,\mu\pm\delta\mu(x))$ in this case.

Next, we consider the low-temperature regime in which $\tau_{\rm ene}(\epsilon)$ is much longer than $\tau_{\rm sf}$ for $\Delta \le |\epsilon| \lesssim eV$. Thus, we can neglect the collision-integral term in eq. (90). Solving eq. (90), we obtain

$$f_{\rm L+}(x,\epsilon) = f_0(\epsilon)\;{\rm e}^{-\frac{|x-x_0|}{\lambda_{\rm s}}} \tag{98}$$

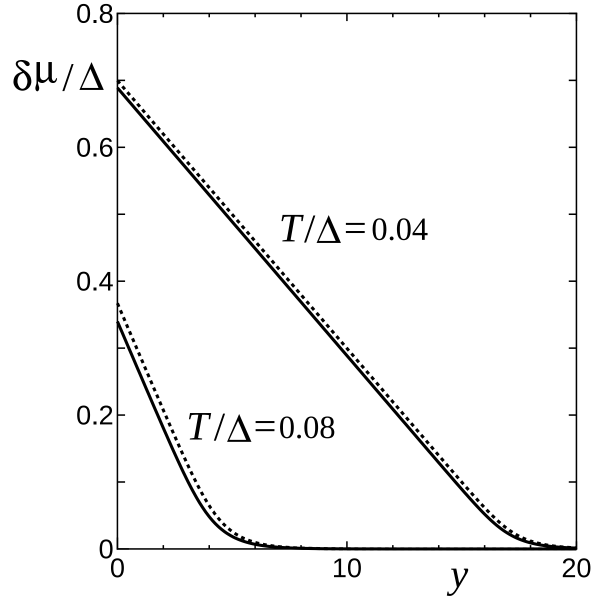


Fig. 1. $\delta\mu/\Delta$ for $T/\Delta = 0.04$ and 0.08 as a function of the normalized distance $y \equiv |x - x_0|/\lambda_{\rm s}$ from the injection point. The solid and dotted lines correspond to $eV/\Delta = 1.2$ and 1.6, respectively.

with

$$f_0(\epsilon) = \frac{\lambda_{\rm s} P_{\rm s}}{16e^2 N_{\rm S}(0) R_t AD} N_1(\epsilon)\left(\tanh(\frac{\epsilon + eV}{2T}) - \tanh(\frac{\epsilon - eV}{2T})\right). \tag{99}$$

In this case, we cannot express the distribution function $f_\sigma(x, \epsilon)$ by the Fermi-Dirac distribution with a shifted chemical potential. Substituting eq. (98) into eq. (89), we obtain

$$\int_0^\infty {\rm d}\epsilon\, N_1(\epsilon)\Big(\tanh(\frac{\epsilon + \delta\mu(x)}{2T}) - \tanh(\frac{\epsilon - \delta\mu(x)}{2T}) - 4f_0(\epsilon)\, {\rm e}^{-\frac{|x-x_0|}{\lambda_{\rm s}}}\Big) = 0. \tag{100}$$

We numerically solve eq. (100) and obtain $\delta\mu(x)/\Delta$ as a function of the normalized distance $y \equiv |x - x_0|/\lambda$s from the injection point $x_0$ at $eV/\Delta = 1.2$ and 1.6 for $T/\Delta = 0.04$, 0.08 and 0.16. The following parameters are adopted: $R_t = 2\ {\rm k\Omega}$, $N_{\rm S}(0) = 1.2 \times 10^{22}\ {\rm eV^{-1}cm^{-3}}$, $D = 5.3 \times 10^9\ \mu{\rm m^2/s}$, $\tau_{\rm sf} = 200$ ps and $A = 50 \times 250\ {\rm nm^2}$. The values of $D$ and $\tau_{\rm sf}$ result in $\lambda_{\rm s} = 1.03\ \mu$m. Figure 1 shows that the bias-voltage dependence of $\delta\mu$ is very weak, in contrast to the high-temperature regime where $\delta\mu \propto eV$. This clearly indicates a nonlinear nature of eq. (100). From Fig. 2, we observe that the decay of $\delta\mu(x)$ can be fitted by the exponential law ${\rm e}^{-|x-x_0|/\lambda_{\rm s}}$ in the case of $T/\Delta = 0.16$. However, except for this case, a deviation from the exponential law appears near the injection point (i.e., $y = 0$). We also observe that the asymptotic behavior of $\delta\mu(x)$ far from the injection point is still governed by the exponential law. It should be noted here that the anomalous slow decay of $\delta\mu$ observed near the injection point is partly attributed to the divergence of $N_1(\epsilon)$ at the gap edge. Since the divergence is smeared by a gap anisotropy, the anomalous behavior may be weakened in actual cases.

Yamashita *et al.*[15] have studied the spin-imbalance relaxation in superconductors by as-

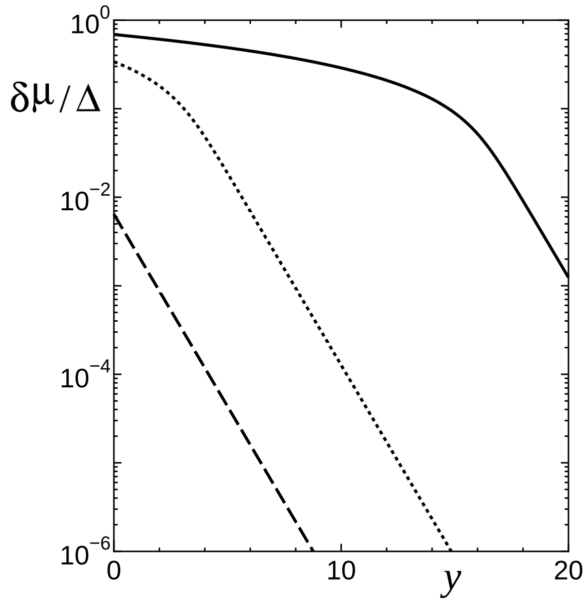


Fig. 2. $\delta\mu/\Delta$ at $eV/\Delta = 1.2$ as a function of the normalized distance $y \equiv |x - x_0|/\lambda_{\rm s}$ from the injection point. The solid, dotted and dashed lines correspond to $T/\Delta = 0.04$, 0.08 and 0.16, respectively.

suming that the quasiparticle distribution function can be expressed in the form of the Fermi-Dirac distribution function with a shifted chemical potential $\mu_\sigma(x) = \mu \pm \delta\mu(x)$, and concluded that $\delta\mu_\sigma(x)$ decays exponentially on the length scale of $\lambda_{\rm s}$. Our argument indicates that the assumption employed in ref. 15 can be justified only in the high-temperature regime. Indeed, near the injection point at low temperatures, we have found deviations from the exponential law.

We have neglected the charge imbalance in the above argument. The nonlinear equation, eq. (89), is reduced to

$$\delta\mu_{\uparrow,\downarrow}(x) = 4T\cosh^2\big(\frac{\epsilon}{2T}\big)\big(\pm f_{\rm L+}(x,\epsilon) + f_{\rm T+}(x,\epsilon)\big) \tag{101}$$

in the high-temperature regime. Thus, an additional spin-independent correction is simply added to the chemical potential if we take the charge imbalance into account. However, such a simple treatment cannot be applied to the low-temperature regime because we must solve eq. (89) in its present form to obtain $\delta\mu_\sigma$. Thus, the spin-imbalance and charge-imbalance corrections to $\delta\mu_\sigma$ are not necessarily additive.

## 5. Summary

We have studied a nonequilibrium distribution of quasiparticles in the presence of both spin and charge imbalances. By extending the kinetic equation approach by Schmid and Schön based on the quasiclassical Green's function method, we have presented a set of linearized Boltzmann equations for distribution functions $f_{\rm L\pm}(\boldsymbol{r},\epsilon)$ and $f_{\rm T\pm}(\boldsymbol{r},\epsilon)$ in steady states. It is

shown that $f_{\mathrm{L}+}$ and $f_{\mathrm{T}-}$ represent the spin and charge imbalances, respectively, and $f_{\mathrm{L}-}$ and $f_{\mathrm{T}+}$ represent the excess quasiparticle energy and the energy imbalance between up- and down-spin quasiparticles, respectively. It is also shown that the suppression of the pair potential due to a current injection is governed by $f_{\mathrm{L}-}$. These distribution functions are decoupled with each other in the Boltzmann equations. This allows us to separately consider the spin and charge imbalances in steady states.

As an application of the Boltzmann equations, we have considered the relaxation of spin imbalance in a quasi-one-dimensional superconducting wire weakly coupled with a ferromagnetic electrode. The spin imbalance induces a shift $\delta\mu$ ($-\delta\mu$) of the chemical potential for up-spin (down-spin) quasiparticles. We have analyzed spatial decay of $\delta\mu$ due to spin-orbit impurity scattering. We have shown that at high temperatures, $\delta\mu$ decays exponentially on the length scale of $\lambda_{\mathrm{s}}$, where $\lambda_{\mathrm{s}}$ is the spin diffusion length. However, at low temperatures, we have observed deviations from the exponential law near the injection point.

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